Quantum criticality of $Ce_{1-x}La_xRu_2Si_2$: the magnetically ordered phase

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We report specific heat and neutron scattering experiments performed on the system $Ce_{1-x}La_xRu_2Si_2$ on the magnetic side of its quantum critical phase diagram. The Kondo temperature does not vanish at the quantum phase transition and elastic scattering indicates a gradual localisation of the magnetism when x increases in the ordered phase.

One important issue in the study of quantum criticality in itinerant electron systems is the determination of the dynamical spin susceptibility and of its relation to the anomalous bulk properties observed near the quantum critical point (QCP). To this aim, the description of 3d itinerant electron systems was achieved by the spin fluctuation theory of Moriya. In 4f electron systems, the occurence of the Kondo effect with a related energy scale of order 10 K leads to a more complex problem. Basically, two scenarii emerge for these systems: the conventional spin fluctuation scenario in the line of Moriya's theory [1,2,3] and the local scenario [4]. Their respective description of the intrinsic nature of quantum criticality deeply differ. In the spin-density wave scenario, the fluctuations of the order parameter, i.e. at *the wave-vector of ordering*, are leading to the transition as in a classical phase transition. A difference with a finite temperature phase transition is that the effective dimension of the system is increased due to quantum fluctuations [2,3]. In this theory, the Kondo effect evolves smoothly accross the QCP. On the contrary, in the so-called local scenario, the low frequency spin dynamics is critical $everywhere$ in the Brillouin zone. In this model, this is associated with a destruction of the Kondo effect at the QCP.

Over several decades, numerous neutron scattering experiments were performed on the archetypal Pauli paramagnet heavy fermion compound $CeRu_2Si_2$ for various dopings leading to long range magnetic ordering [5,6,7,8]. For $Ce_{1-x}La_xRu_2Si_2$, sine-wave modulated magnetic ordering occurs for $x \geq x_c$=0.075 at the incommensurate wavevector $\mathbf{k_1}$=(0.31, 0, 0), the magnetic moments being aligned along the c-axis of the tetragonal structure, due to the strong Ising anisotropy originating from the crystal field. In the present paper, we report recent data taken in the magnetically ordered phase for x=0.13 (T_N=4.4 K) and x=0.2 (T_N=5.8 K) that extends the works performed in the past for x=0.13 [9], x=0.2 [10,11] and x=0.3 [12].

Figure 1 shows the magnetic specific heat divided by the temperature for several concentrations accross the critical point (data for x=0 and x=0.075 are taken from Ref.[5]). While focus is usually made on the critical concentration, the most striking features occur in the ordered phase. For x=0.13, C_m/T is almost constant in the ordered phase. For x=0.2, the slow decrease of C_m/T below T_N is amplified below a second transition temperature T_L = 1.8 K. For x=0.13, the low transition occurs at T_L $\approx$ 600 mK. We note that the decrease of C_m/T below T_L for x=0.13 is far smaller than the one reported earlier [14]. The huge effective mass related to the high value of C_m/T for T $\rightarrow$ 0 at x=0.13 already suggests that itinerant magnetism is characteristic of the ordered phase. The anomalous shape of the x=0.13 specific heat curve can be qualitatively understood as a the sum of a large spin fluctuation contribution and of a specific heat jump related

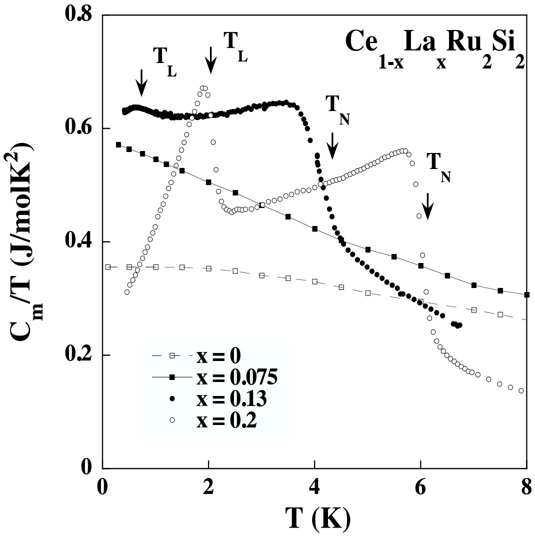

Figure 1 Magnetic specific heat of $Ce_{1-x}La_xRu_2Si_2$ divided by temperature for x=0, 0.075, 0.13 and 0.2.

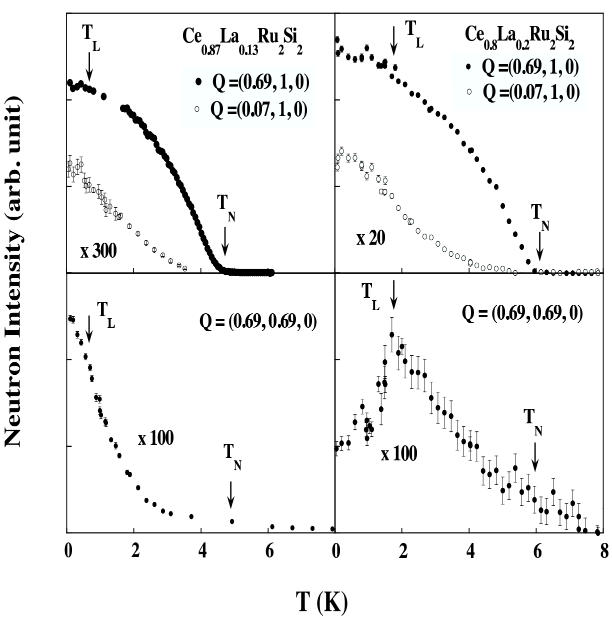

Figure 2 Temperature dependence of the elastic scattering measured at $\mathbf{Q}$=(0.69, 1, 0), $\mathbf{Q}$=(0.07, 1, 0) and $\mathbf{Q}$=(0.69, 0.69, 0) for x=0.13 (left) and x=0.2 (right).

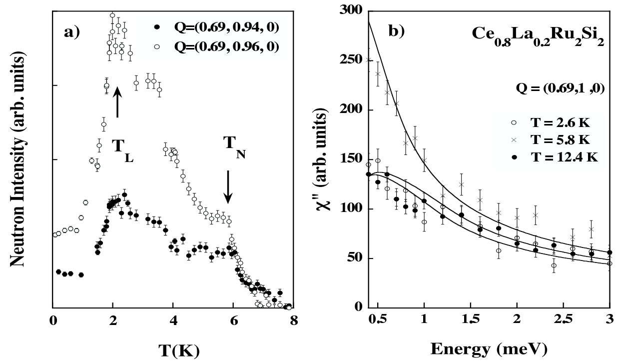

Figure 3 a) Temperature dependence of the diffuse scattering measured at $\mathbf{Q}$=(0.69, 0.96, 0) and $\mathbf{Q}$=(0.69, 0.94, 0) for x=0.2. b) Energy spectra measured for $\mathbf{Q}$=(0.69, 1, 0) at 2.6, 5.8 and 12.4 K.

to the magnetic phase transition. To better understand the magnetic state of $Ce_{1-x}La_xRu_2Si_2$, neutron scattering measurements were undertaken for x=0.13 and x=0.2 on the cold three axis spectrometer IN12 at the Institut Laue Langevin, Grenoble. Details of the inelastic measurements are given elsewhere [13].

The temperature dependence of the elastic intensity is shown in figure 2 for the wave-vector of ordering $\mathbf{k_1}$=(0.31, 0, 0) (measured at the momentum transfer $\mathbf{Q_1}$=(0.69, 1, 0)=(1, 1, 0)-$\mathbf{k_1}$), the third harmonic of $\mathbf{k_1}$ (measured at the momentum transfer $\mathbf{Q}$=(0.07, 1, 0)=(1, 1, 0)-3$\mathbf{k_1}$), and for the wavevector $\mathbf{k_2}$=(0.31, 0.31, 0) at which short range magnetic fluctuations are observed in pure $CeRu_2Si_2$ and here conveniently measured at $\mathbf{Q}$=(0.69, 0.69, 0)=(1, 1, 0)-$\mathbf{k_2}$. For x=0.13, the third harmonics is barelly developped with $I_{\mathbf{k_1}}/I_{\mathbf{3k_1}} \approx 500$, while fluctuations at $\mathbf{k_2}$ are substantial with $I_{\mathbf{k_1}}/I_{\mathbf{k_2}} \approx 100$ ($I_{\mathbf{k}}$ is the elastic intensity at the wavevector $\mathbf{k}$). For x=0.2, the elastic scattering at $\mathbf{k_2}$ is maximum at T_L and $I_{\mathbf{k_1}}/I_{\mathbf{k_2}} \approx 300$ at 50 mK. Similar maxima of the elastic scattering at T_L are observed at wave-vectors in the vicinity of $\mathbf{k_1}$. This is shown in figure 3a for $\mathbf{Q}$=(0.69, 0.96, 0) and $\mathbf{Q}$=(0.69, 0.94, 0). For these wave-vectors "in the foot" of the $\mathbf{k_1}$ peak (the corresponding full width at half maximum of the magnetic Bragg peak being 0.04 r.l.u.), a maximum of the diffuse scattering is expected at T_N for a classical finite temperature phase transition. Such diffuse scattering in the vicinity of $\mathbf{k_1}$ is not observed for x=0.13. Concerning the third harmonics, we obtained $I_{\mathbf{k_1}}/I_{\mathbf{3k_1}} \approx 40$ for x=0.2. This increase of the third harmonics from x=0.13 to x=0.2 points out to an increase of the local nature of the magnetism. Indeed it corresponds to a stronger squaring of the modulated structure and consequently to a decrease of the longitudinal fluctuations ($I_{\mathbf{k_1}}/I_{\mathbf{3k_1}}$=9 for a squared modulation). We stress on the fact that the nature of the magnetic ordering below T_L is still unknown. Current hypotheses correspond to a lock-in of the phase of the modulation [10] or to a transition from a single-$\mathbf{k}$ structure to a double-$\mathbf{k}$ structure [15]. Figure 3b shows typical energy spectra obtained for x=0.2 at the constant momentum transfer $\mathbf{Q}$=(0.69, 1, 0) corresponding to the wave-vector $\mathbf{k_1}$ for T=2.6, 5.8 and 12.4 K (See Ref.[13] for more details). Such spectra were analyzed using a Lorentzian lineshape characterized by a single linewidth corresponding to a wave-vector dependent relaxation rate. Figure 4 shows the temperature dependence of the magnetic relax-

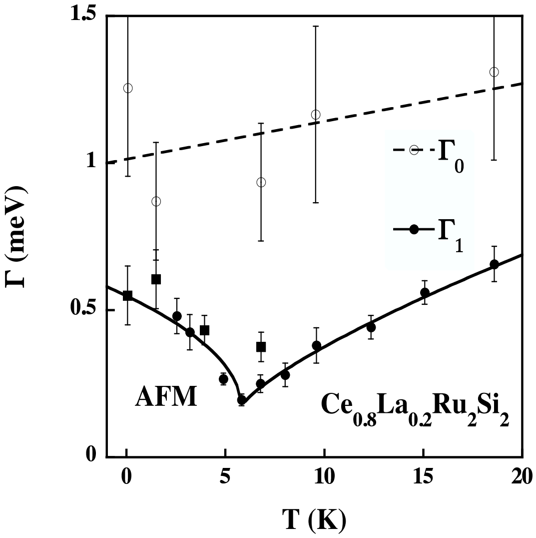

Figure 4 Local (open symbols) and antiferromagnetic (full symbols) relaxation rate of $Ce_{0.8}La_{0.2}Ru_2Si_2$ as a function of temperature. Full circles correspond to measurements performed at **Q**=(0.69, 1, 0) while full squares correspond to measurements performed for **Q**=(0.72, 1, 0) i.e. slightly off the $\mathbf{k_1}$ peak position. The local relaxation was measured at **Q**=(0.44, 1, 0).

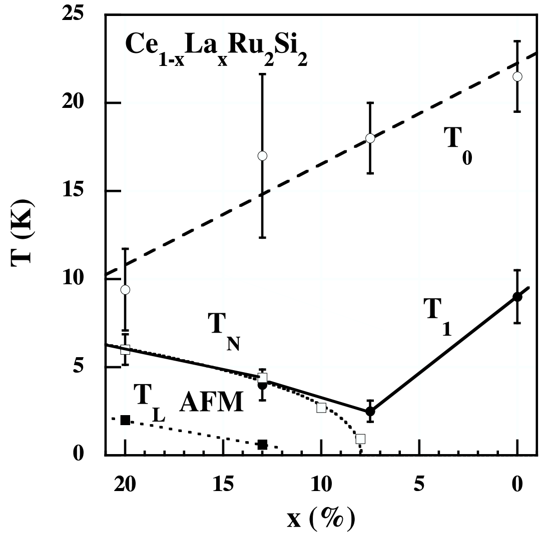

Figure 5 Magnetic phase diagram of $Ce_{1-x}La_xRu_2Si_2$ obtained by elastic and inelastic neutron scattering [13].

ation rates measured by inelastic neutron scattering for x=0.2 at two wave-vectors [13]. At the wave-vector $\mathbf{k_1}$ of magnetic ordering, the relaxation rate Γ_1 is minimum at T_N. However its value is finite as for the value of the relaxation rate at the critical concentration x_c for T $\rightarrow$ 0 [6] (See below). The relaxation rate Γ_0 corresponding to fluctuations measured far from the ordering vector, here at the momentum transfer **Q**=(0.44, 1, 0), does not show any anomaly at T_N. The persistence of local relaxation inside the magnetically ordered phase is interpreted in terms of a finite Kondo temperature at $T \rightarrow 0$ for x=0.2. Very similar data were also obtained for x=0.13 [13].

Figure 5 summarizes the quantum critical phase diagram obtained by neutron scattering measurements [13]. T_1 is the extrapolation of the relaxation rate Γ_1 for $T \rightarrow 0$ ($k_B T_1 = \Gamma_1(T)_{T\rightarrow 0}$) and T_0 is the extrapolation of Γ_0 for $T \rightarrow 0$ ($k_B T_0 = \Gamma_0(T)_{T\rightarrow 0}$). T_1 shows a minimum at x_c and is finite [6]. T_0 does not show any anomaly at x_c, which evidences again that the Kondo fluctuations are finite in the magnetically ordered phase. It is worthwhile to note that T_1=T_N in the ordered phase. Figures 4 and 5 show that the behavior at $T \rightarrow 0$ as a function of x is similar that the one for x =0.2 as a function of T. The minimum of T_1 at x_c (or of Γ_1 at T_N) and the absence of anomaly of T_0 at x_c (or of Γ_0 at T_N) suggest that the fluctuations at the wave-vector $\mathbf{k_1}$ are responsible for both the classical and quantum phase transitions. The finite value of the characteristic energy at T_N or x_c suggests a first order character of the phase transition.

To conclude, our data confirm that the quantum critical point of $Ce_{1-x}La_xRu_2Si_2$ is of itinerant nature. This is evidenced by the finite value of the Kondo temperature in the ordered phase and by the associated absence of anomaly in the local relaxation rate at x_c or T_N. In the magnetically ordered phase, the magnetism gradually evolves from itinerant to localized, as evidence by the increase of the squaring of the modulation and by the maximum of diffuse scattering occurring at the finite temperature T_L for x=0.2 but absent for x=0.13. The fact that it occurs at T_L rather than at T_N as expected is unclear and would merit further investigation, in relation with the question of the nature of the second transition at T_L.

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